CHAPTER 3

GEOLOGICAL RADWASTE DISPOSAL IN BELGIUM RESEARCH PROGRAMME, REVIEW AND OBJECTIVES

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Executive Summary. In the early seventies, SCK•CEN launched a research programme on the geological disposal of long-lived radioactive waste. Investigations were concentrated on argillaceous formations because of their abundance at various depths in some parts of Belgium, their expected favourable properties and the virtual absence of salt and/or unfracturated hard rock formations in the country. Among the argillaceous formations, the Tertiary "Boom clay" layer was tentatively selected for site characterization studies. In this first phase, the Research and Development programme consisted of laboratory studies on clay core samples and observations on the geology and hydrology of the area. The initial results were very promising with regard to sorption properties of the potential host rock, its chemical stability, permeability, etc.

Early in the programme (1980), an underground laboratory (HADES) was constructed in the clay at the Mol site; a tunnel, 35 m long and 3.5 m in diameter provides a large number of access possibilities to the clay formation, at a depth of 223 m. The first objectives were related to the technical feasibility of gallery excavations in plastic clay under high lithostatic pressures and to the evaluation of the compatibility of vitrified HLW and candidate container materials with the clay, in close to real conditions. In 1988, an additional 60 m long drift was built for integrated tests, e.g. the impact of temperature and radiation. In the meantime, much experience has been gained on sampling of fluids and solids, in the characterization of clay and waste forms; an important scientific data base is now available at SCK•CEN, specifically for vitrified waste and "Boom clay."

This formation - and site specific approach - allowed SCK•CEN to collect, in a short period of time, valuable results and input data for required modelling work. Some preliminary observations could be made:

- using conventional civil engineering technology, it is technically feasible to drive horizontal galleries in plastic clay, at a depth of more than 200 m, with an internal diameter of at least 3.5m, without special ground conditioning, such as freezing;
- using available experimental data about permeability and retention properties of the host rock (Boom clay), preliminary safety assessments were carried out for many scenarios from natural evolution to important disruption. For the reference mix of reprocessing waste (HLW, MLW, hulls) from a 40 y nuclear programme in Belgium, the highest calculated dose to population, for a normal evolution scenario and water well pathway, is of the order of 3×10^{-7} Sv/a, which can be compared with a dose constraint of 10^{-4} Sv/a, in use in several countries; these doses would occur after a period of about 60,000 years.
- stabilizing factors in the Boom clay are its cationic exchange capacity, the strong reducing conditions due to finely dispersed pyrite and humic materials intimately associated with the clay minerals and a slightly alkaline environment caused by the occurrence of carbonates; and
- the methodology for risk assessment was established and tested in the framework of an international benchmark exercise in cooperation with the European Commission and the Nuclear Energy Agency of the OECD.

Observations and earlier assessments also illustrate remaining areas of uncertainty which require complementary R&D or new studies and demonstrations. Furthermore, the validity of present evaluations and preliminary conclusions, mainly in view of the very long term safety, will benefit from an iterated validation of

models and even comparisons with studies on natural analogues.

3.1 INTRODUCTION

In the present context, the R&D programme of SCK•CEN on radioactive waste management is focused on the characterization of conditioned waste and its underground disposal in geological formations.

Although in the past (1960 to ~1985), SCK•CEN has run several projects on waste treatment and conditioning (mainly low-level and alpha-bearing waste), very little is still being pursued in this area, given the commercial availability of industrial processes and the fact that the waste treatment and conditioning has been entrusted to a separate company, BELGOPROCESS. However, this trend may have to be adjusted some time in the future for reasons of optimization and the development of new waste types resulting from e.g decommissioning and/or site restoration programmes.

With regard to characterization of conditioned waste, the programme aims at quality control (verification of compliance with contractual quality criteria), characterization of the "source term," which includes studies of corrosion phenomena and compatibility of the conditioned waste form, and its package with the intended geological environment. Attention also has to be paid to "acceptance criteria" since the total radiological capacity of the repository is obviously limited, and non-radiological factors (e.g temperature and residual thermal capacity of the waste and potentially interfering components) may enhance the mobility of radionuclides in the repository formation. Furthermore, the occurrence of chemo-toxic components in conditioned waste or packages deserves a minimum of attention.

With regard to geological disposal, the R&D programme, initiated by SCK·CEN, deals mainly with the disposal of vitrified high-level waste from reprocessing and other alpha-bearing wastes. Other types of waste are to be considered in the future such as, for example, non reprocessed spent fuel or spent MOX fuel since it is clear that, even if reprocessing remains the reference scenario, at least some spent fuel is not suited for industrial reprocessing.

The R&D programme on disposal proceeds stepwise and cautiously since options have to remain flexible until adequate scientific and technical ground is available to support decisions.

In the exploration of geological formations (1974) for the implantation of a repository in Belgium and after consultation with the National Geological Survey, preference was expressed for a deep clay formation (called "Boom clay") at and around the site of Mol. This early preference, resulted from a number of facts, e.g. the absence in Belgium of any salt formations, the highly fractured nature of most hard rock formations, the natural retention capacity of clay, and the fact that major nuclear facilities were already installed in the Mol area. Consequently, R&D and assessment studies became mostly site-specific or at least formation-specific, without anticipating the conclusions of a more detailed site selection procedure.

The Boom clay belongs to the Rupelian formation and is underlain by the Berg sands (Rupelian aquifer) and overlain by the Neogene aquifer. These two water bearing formations are not considered as barriers, although they could have good sorption properties for the radionuclides. Rather early in the programme (1980), an underground laboratory was constructed in the clay at the Mol site at a depth of 223 m. The underground laboratory was intended to evaluate the technical feasibility of such a construction and to become an *in-situ* facility for performing tests in close to real conditions within the High Activity Disposal Experimental Site (HADES) project.

The current R&D programme is focused on site characterization (mechanical, physicochemical, and hydrogeologic properties) and performance assessments. Understanding the basic phenomena which control the retention and/or mobility of radionuclides in the clay received high priority. A first interim safety report (SAFIR) was produced for the Authorities in 1989. The main conclusion was the absence of counter-indications against the principle of disposal in clay. The next preliminary safety report is to be submitted around 1998 and is meant to be an important milestone in the selection, or decision making, process.

For the near future, investigations on basic phenomena of physical chemistry and geohydrology are to be continued; in addition, a number of areas may require much more study and investigation such as:

- impact of radiation and heat on retention/mobility of radionuclides in clay; these near-field effects may affect important factors such as the length of cooling time in surface storage facilities prior to transfer into the repository and/or the conceptual design of the repository;

- influence or significance of non-radioactive components in the anaerobic environment of the repository (e.g decomposition products of organic materials). In case of adverse effects, they might lead to the consideration of alternative conditioning processes, depending on the acceptance criteria for packaged waste; and
- other issues are to be considered as well, e.g position with regard to retrievability, reduction of uncertainties with regard to some major characteristics and to the time scales applicable to geological disposal.

The above programme items require a major extension of the present underground facility to perform larger scale integrated *in-situ* demonstration tests. This important decision has been recently taken and after 1998 will lead to doubling the present capacity.

3.2 THE HADES PROGRAMME AT SCK·CEN

The R&D programme on geological disposal was initiated in 1974 by SCK•CEN and received scientific and financial support from the European Commission from the very beginning. Since 1985, it has been heavily supported by the National Radioactive Waste Agency (ONDRAF/NIRAS), which is in charge of the implementation of the disposal programme.

With a total installed nuclear power of 5.5 GWe and a 40-year operational period for the Belgian nuclear power plants, the volume of waste to be produced of low, medium and high activity can be estimated to be 150,000 m^3, 39,000 m^3 and 4000 m^3, respectively. These volumes do not yet include the decommissioning waste.

The first category of waste contains small quantities of short- or medium-lived radionuclides (half-life not exceeding thirty years); the current option is to dispose this waste at the surface; shallow land burial is considered in Belgium as the reference solution but will not be examined in the present document. Earlier disposal studies resulted in a report submitted by the waste management authority (NIRAS/ONDRAF) in 1993 to a national advisory scientific commission; they are being updated and extended according to the recommendations of this commission.

Two other types of waste are considered in Belgium for underground disposal:

- alpha-contaminated waste (concrete or bitumen matrix) containing significant quantities of medium-lived radionuclides (half-life exceeding 30 years); it results from specific fuel cycle operations (spent fuel reprocessing, fuel assembly manufacturing) and does not generate heat; and
- vitrified heat-emitting waste (glass matrix) containing a mixture of short- or medium-lived radionuclides in high concentrations, and long-lived radionuclides which are usually alpha emitters. It generates a significant heat power, owing to the fission products content.

Table 3.1 presents some more details on these waste types and volumes.

Table 3.1. Data on radioactive waste types in Belgium.

Waste Type	Matrix	Volume Total (m^3)
HEAT EMITTING WASTE		
Cogema high-level waste	glass	784
Cogema cladding waste	concrete	2,860
Eurochemic high-level waste	glass	265
	Total	3,909
ALPHAWASTE & OTHER		
Technological alpha waste	cement	8,419
MLW concentrates	concrete	20,430
Reactor operating waste	concrete	2,724
Coprecipitation sludge	bitumen	3,310
Eurochemic MLW concentrates	bitumen	2,765
Mox production waste	concrete	1,353
	Total	39,001

It is assumed in the present reference scenario that the operation of a repository for underground waste disposal could start in 2035 to last until 2070/2080 (closure phase). This requires, for vitrified waste, a temporary surface storage of about 50 years allowing the heat output to be significantly reduced. This limited heat output in the repository is intended to produce temperatures not exceeding 100° C in the host clay at the container/clay interface and is one of the basic assumptions for the definition of the R&D programme and associated processes to be considered.

3.2.1 Historical Review of Hades Program

Early studies on potential host formations for geological disposal have indicated that, in Belgium, clay and shale layers can be taken seriously into consideration. One of the potential formations which was identified is the Boom clay at the Mol-Dessel site situated in northeastern Belgium, where the SCK•CEN and other nuclear facilities are located. The following guidelines for the provisional selection (for experimental purposes) of the Boom clay layer were considered: the anticipated future stability of this 30 million year old formation as well as its depth, thickness, relative homogeneity and sorption/retention capacity (Heremans and Baetslé, 1978).

The research programme was started about 20 years ago by SCK•CEN, which concentrated its activities on verification of basic phenomena, safety and technical feasibility. Several partners from Europe and abroad joined the programme during the second decade and, through their own competence, contributed to the integration of different scientific and technological skills and approaches (Bonne and Collard, 1992).

3.2.2 Approach and MajorAchievements During First Two Decades of Programme.

Beside the selection of the formation (and experimental site), the first decade (1974-1984) was dedicated to verify whether the characteristics and properties of the Boom clay layer were really promising for hosting a geological repository.

The first drillings were performed on the SCK•CEN site from 1975 onwards; and analyses of core samples were made to determine lithological, chemical, mineralogical, ion exchange and geomechanical properties of Boom clay and, in some cases, of the surrounding strata. Geohydrological studies were undertaken; a preliminary repository design and a probabilistic risk assessment methodology were developed. Simultaneously, a catalogue of all potential formations occurring in the country was established. From these studies, it was concluded that the Boom clay satisfied the expectations; this plastic material has good sorption capacities and provides very low permeability and low but sufficient heat conductance. It is sufficiently thick and homogeneous and it is chemically and mineralogically stable. Preliminary experiments on corrosion/heat transfer were performed in parallel from an open clay pit in Terhaegen, near Antwerp.

The next step was to demonstrate that it was technically feasible to build a repository in such a plastic material and to strengthen our capabilities in the area of safety assessment; in the latter, a collaboration with JRC Ispra was an important feature (D'Alessandro and Bonne, 1981).

An underground research laboratory (URL) was built according to specifications which were based on very pessimistic hypotheses (afterwards found to be unrealistic) about the plasticity and perviousness of the Boom clay. For example, the freezing technique was not only used in sands to sink the shaft through these water-bearing formations but also in clay to lower the anticipated convergence rate.

The approach became fully site and formation specific; the site of Mol was placed in a larger geohydrological context, extending the regional observation network on a 2,000 km^2 area around Mol. Both clay and surrounding geologies were intensively investigated from various perspectives (geochemical, geomechanical, hydrological and geophysical). Due to an increasing amount of available information, the Boom clay became progressively a reference case.

For the critical issue of geomechanics in clay, collaboration was started with ANDRA, the French waste management agency; the digging of a small experimental shaft and gallery in non-frozen clay demonstrated the possibility of excavating and tunneling in the plastic Boom clay at such a depth.

During the second decade (1984-1994), the URL became fully operational; several experimental set-ups were installed in clay for studies on corrosion, hydraulics, migration and geomechanics. A new and challenging field of demonstration was opened with the CEC programme on demonstration and pilot facilities. The HADES URLwas extended by tunneling a test drift for demonstration purposes as illustrated in Figure 3.1.

This facility, now called URF (Underground Research Facility,) consists of 100 metres of galleries lined with cast-iron, concrete or steel ribs, providing direct access to the clay mass. In particular, the test drift offered the opportunity to perform large scale integrated tests under conditions close to those expected in and around a final repository.

Validation exercises for the modelling of different processes were launched and extensive performance assessment exercises were carried out, as the "Performance Assessment of Geological Isolation Systems" (PAGIS) and the "Performance Assessment of

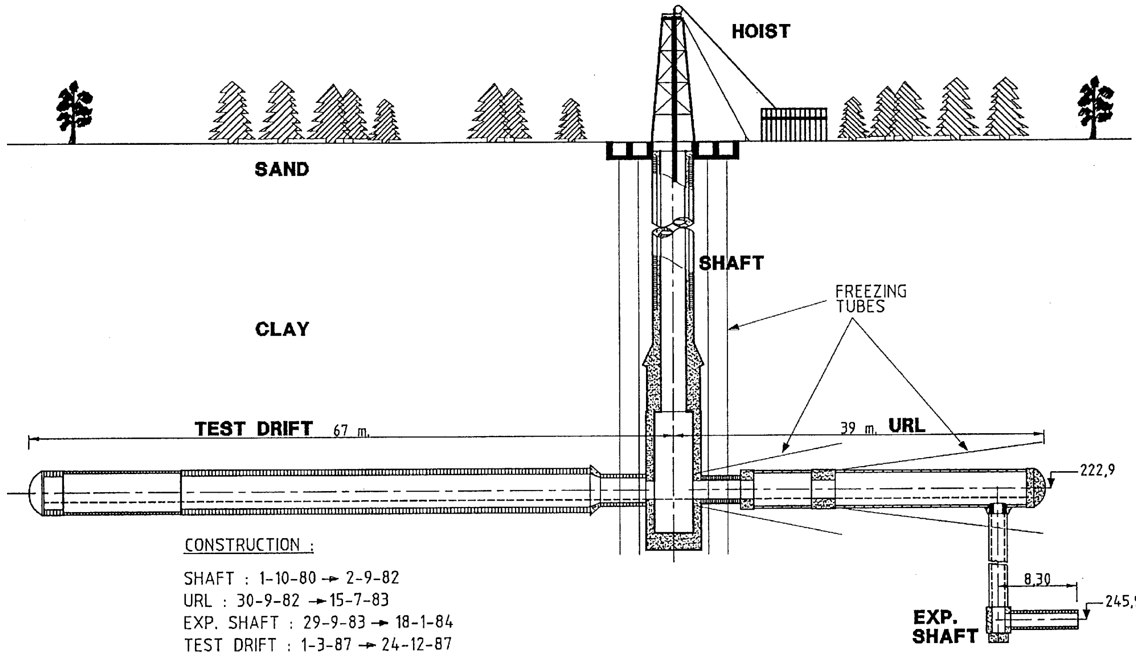

Figure 3.1. The underground research facility (URF) HADES.

Confinements for Medium-level and Alpha waste," (PACOMA) within the framework of the E.C. Both studies were updated, and results were published in the "Updating 1990" (Marivoet, 1992). Amethodology was elaborated with the involvement and consensus of a large number of scientists in different member states, active in the various fields of this multidisciplinary activity; the broad contribution from the latter activity had a very positive impact on confidence building.

In 1990, an international commission of experts, formed by the Belgian Secretary of State for Energy, evaluated a Safety Assessment and Feasibility Interim Report (SAFIR) on the geological disposal of waste prepared by NIRAS/ONDRAF (1989); the results of the research carried out by SCK•CEN on final disposal in Boom clay were the main contribution to this report.

These successive actions confirmed the choice of the clay option in Belgium and the continuation of the site characterization programme on the Boom clay layer at the Mol-Dessel site as a potential host formation essentially for long-lived waste. They also define the R&D priorities developed in the current programmes on the different components of the disposal system. As a result of these continuing interactions, tangible progress has been made through improvements in the SCK's characterization programme, e.g. more recently regarding the study of the performance of the engineered barriers in the near-field. Present and future developments are detailed in the next chapters.

3.3 OBJECTIVES AND BASIC ASSUMPTIONS OF HADES PROGRAMME

3.3.1 Objectives

Safe disposal of radioactive waste is one of the key issues in the environmental concerns of the nuclear industry. In particular, the disposal of long-lived waste, specifically spent fuel and vitrified high-level wastes, is important. Disposal in deep geological formations is at present the most promising option, which requires one to meet the following objectives:

- provide reliable data on characteristics of waste and the geological environment;
- demonstrate the feasibility of disposal concepts e.g. in underground laboratories; and
- assess the safety and acceptability of disposal systems by developing and applying validated methods to the modelling of the various phenomena which control the release and migration of radionuclides from the repository into the biosphere.

3.3.2 Methodology

In order to assess the performance of individual components of the disposal system as well as that of the integrated system, the current R&D programme covers mainly research on basic phenomena, demonstration tests and safety studies.

The geological disposal of long-lived waste is essentially based on a “multi-barrier” concept; several “engineered” barriers (overpack, backfill, gallery lining) are installed between the waste matrix itself (primary containment) and the host rock. The purpose is to delay (e.g. by 500 to 1000 years) the release of activity from the repository structure into the geological environment. The latter time period corresponds approximately to the thermal phase of the repository. Consequently, several coupled effects with temperature can be avoided in the host clay, provided the required performance of these engineered barriers can be guaranteed.

Phenomena relevant to repository performance are generally subdivided into those occurring in the near-field and those occurring in the far-field (geosphere). By near-field, one generally considers the engineered repository structure (including the waste packages and engineered barriers) and those parts of the surrounding rock whose characteristics have been, or could be, altered by the repository or its content (excavation, temperature, radiation).

Any assessment of the performance of geological disposal refers to a conceptual design of the repository and, in particular, to the characteristics of the near field. Information concerning the Belgian reference concept is described in a separate section (see Section 3.3.4).

3.3.3 Performance of Near-Field

An essential distinction for disposal in clay, with regard to disposal in granite, consists of the requirements and objectives of the near-field.

In hard rock, the components of the multi-barrier containment and essentially the backfill material are to be considered as primary barriers which ensure the isolation of the system for the time periods required. In this context, the backfill must preserve alkaline conditions during a very long time period in order to prevent dissolution of key radionuclides and to provide sorption capacity and controlled porosity and permeability. The host geological formation is supposed to provide structural stability, a predictable groundwater flow, and a suitable long pathway and travel time for radionuclides to reach the biosphere.

For argillaceous formations and particularly for the Boom clay formation, the host “rock” is the primary natural barrier against radionuclide migration; it provides good sorption capacities, very low permeability, and given the composition of the clay, chemically reducing conditions. The backfill has to provide geomechanical and geochemical stability and compatibility.

3.3.4 Repository Concepts in Belgium.

We are considering for geological disposal the long-lived medium level waste (MLW), heat producing vitrified high-level waste (HLW) and spent fuel (SF), but for the latter category, no detailed concept has yet been worked out.

Since 1978, different repository concepts have been considered for high-level and long lived radioactive waste in the Boom clay formation at Mol. The reference concept, now considered by NIRAS/ONDRAF, is illustrated in Figure 3.2. Heat producing high-level and long-lived, medium-level waste would be emplaced in separate disposal galleries. Approximate dimensions (expressed in terms of the inner diameter) for the different components are 6 m for the shafts, 4 m for the primary galleries and 2 m for the HLW disposal galleries. The length of the disposal galleries would be 800 m (4 sections of 200 m). Concrete would be used for lining the shaft and galleries. The lining is designed to ensure mechanical stability during the repository’s operational phase.

For the non-heat producing long-lived MLW, the galleries would have an inner diameter of 3.5 m and the section would be filled with MLW canisters, the voids being backfilled, possibly with concrete (see Fig. 3.2).

The design of the HLW disposal galleries is more advanced (see Fig. 3.3) and will, during the next 10 years, be the main subject of the PRACLAY project. The latter requires a substantial extension of the present URF; it is managed by an Economic Interest Group (E.I.G.) where SCK•CEN and NIRAS/ONDRAF are represented. The PRACLAY project is a thermo-mechanical experiment intended to simulate a full scale disposal gallery for heat-emitting waste and to quantify the extent of the disturbance induced in the surrounding clay.

According to this concept, the HLW canisters, simulated by heating elements, are assumed to be placed in long thin metallic tubes in the centre of the disposal galleries. These tubes are designed to remain intact during the

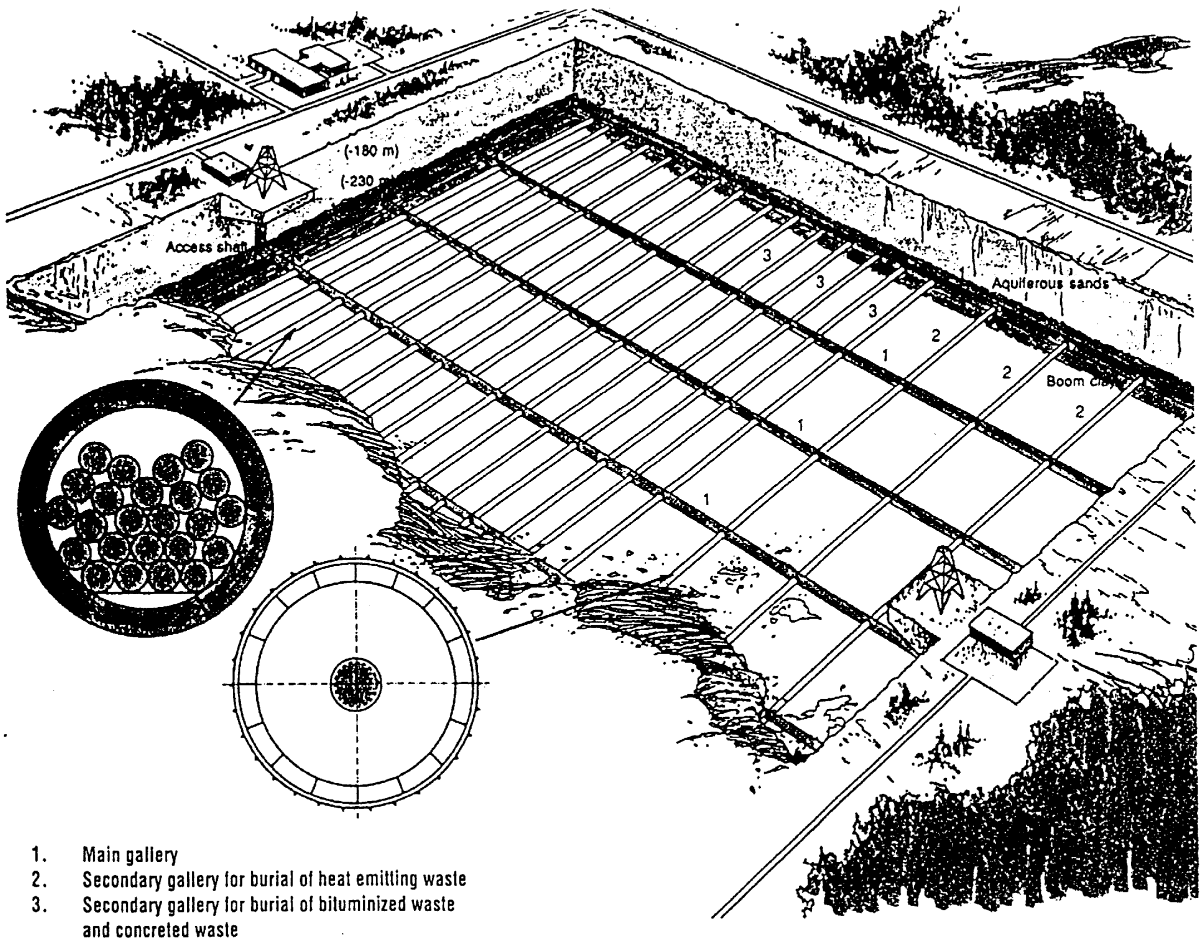

Figure 3.2. Present Belgian reference disposal concept (NIRAS/ONDRAF).

thermal transient of the heat producing waste. The thickness of this "overpack" will have to be sufficient to guarantee its role as a reliable barrier during the thermal phase of the disposal, and sufficiently low to limit the potential generation of gas, through corrosion, in the anaerobic phase. The void between the metallic tube and the concrete gallery lining would, according to the present concept of NIRAS/ONDRAF, be backfilled with pre-compacted calcium bentonite blocks.

The option of retrievability or "long term storage" is examined abroad in some disposal programmes (France, the Netherlands, etc.), possibly in connection with later "advanced reprocessing." However, in Belgium, easy retrievability is not considered, up to now, as a requirement in the different disposal concepts. It is assumed that, with the necessary capital and effort, the retrieval of the waste will always be possible during a limited time period, generally considered to be in the range of 50 to 100 years. Only preliminary studies related to the impact of retrievability on performance assessment might be launched in the near-future by SCK•CEN, providing European partners join the effort.

For the disposal of spent fuel, conceptual studies are in a preliminary stage. The main differences compared to the heat producing HLW are the longer thermal transient to be considered, the content of the total inventory of plutonium and the larger dimensions of the packages. Given the limited dimensions of the clay disposal infrastructure, a size reduction and a form of preconditioning of spent fuel might be required.

3.4 PROGRESS OF CURRENT RESEARCH AND STUDIES

An understanding of the migration processes in the host rock, together with the hydrogeological context of the host and surrounding formations are of prime importance for the long term safety of a reference geological

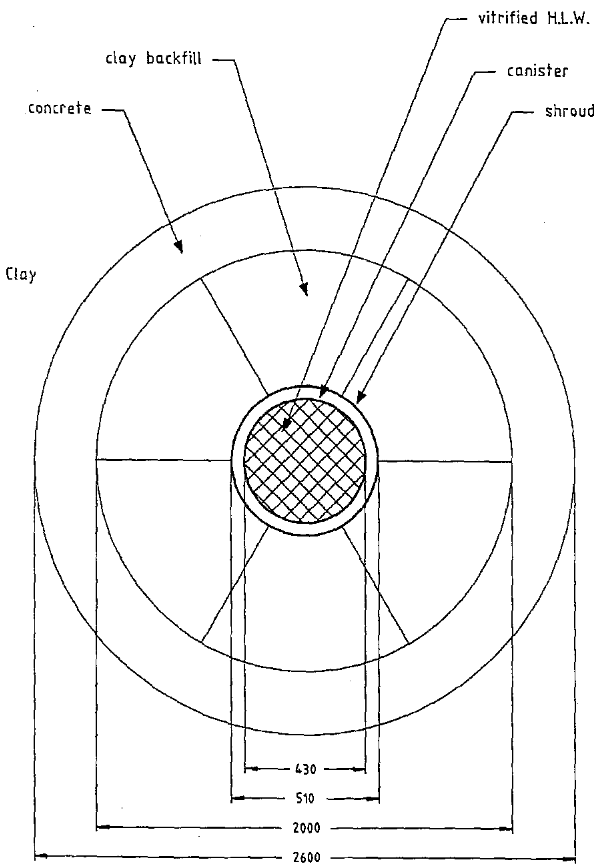

Figure 3.3. Detail of the Belgian reference concept for HLW (NIRAS/ONDRAF).

disposal concept. Furthermore, one has to take into account the technical feasibility of the different components of the system, from the constructional and operational aspects to the assessment of the integrity of the volume of clay surrounding the disposal galleries. All the phenomena and processes involved in and around a deep geological repository (decompression, radiolysis, heat emission, gas production, etc.) and their evolution with time are dependent upon the waste types and the concept of the disposal infrastructure.

3.4.1 Research on Basic Phenomena

The safety of a repository for radioactive waste is conditioned by the long term behaviour of the various elements which constitute the engineered and natural barriers. Research on the basic phenomena expected to take place in the disposal system is therefore essential, in addition to the technical demonstration tests. Some of the research topics listed below are carried out in surface laboratories, others in the underground facility (URF).

This research work is intended to better understand the phenomena, which control the release of radionuclides from the waste packages, and their migration through the various successive barriers to the environment, in order to model these phenomena and validate the models. We are considering the following succession of research issues:

- characterization of waste packages and their compatibility with clay;
- geomechanical/geochemical behaviour of clay-based materials
- gas generation and transport;
- thermal, radiation and chemical effects; and
- radionuclide migration.

3.4.1.1 Characterization of Waste Packages and their Compatibility with Clay

Waste package is a general term used to indicate the conditioned waste in its container; the conditioned waste can be homogenous (vitrified waste, bitumenized sludge) or heterogeneous (e.g supercompacted waste in a cement matrix). “Container” is here considered as a generic term covering both the canister, into which the conditioned waste is loaded, and the overpack which has to provide isolation of the waste for relatively long time periods.

Background

Studies on waste packages performed at the SCK•CEN over the past 15 years have dealt with:

1. characterization of conditioned waste forms, as part of the national QA/QC programme (qualification of the processes, verification - Van Iseghem et al., 1990); and
2. compatibility with the anticipated disposal environment. These studies involved both laboratory and *in situ* “corrosion” tests. The efforts so far mainly focused on glass and bitumen waste forms, C-steel container material and, increasingly, on cement waste forms and stainless steel. These studies were aimed at characterizing the source term and at providing information for the future elaboration of acceptance criteria for the disposal of waste in the host formation.

Results

High-level waste glass (Van Iseghem, 1994; Lemmens et al., 1993). The results of *in situ* experiments are in agreement with those of laboratory tests which show that glass is an efficient first barrier for the radionuclides; dissolution rates of less than 0.1 µm per year were recorded at an ambient clay temperature (16° C) in direct contact with Boom clay. However, the dissolution of glass is strongly enhanced by increasing the temperature and in the presence of clay, which acts as a sink for many glass constituents such as Si, rare earths and actinides. The long-lived actinides (Pu, Am, Np) in general leach slower than the bulk glass, and for more than 90%, the fraction leached is sorbed onto the clay. These data are further used in impact assessment studies reported under Section 3.4.3.

Bitumenized reprocessing waste (Berghman et al., 1990). A high leaching rate of NaNO3 was noticed for Eurobitum, a reference Belgian bitumen, containing an average 35% of reprocessed sludge. By extrapolation, we can predict that a full size drum would be depleted in NaNO3 (25%) within a few thousand years leading to the release of important amounts of nitrates into the near field. Lower leaching rates were noted for the actinides (Pu, Am); their leaching appears to be controlled by solubility and is not enhanced by lithostatic pressures.

Container (Cornelis and Van Iseghem, 1994). An average corrosion rate of 5 µm per year was inferred from *in situ* tests on carbon steel lasting up to 7.5 years; pitting processes with an average growth rate of about 20 µm per year are dominating. Corrosion proceeds mainly by pitting attack in the aerobic phase and congruent corrosion would dominate in the anaerobic phase. Stainless steel is becoming the candidate overpack material.

Future actions

Additional or corrective actions are required due to not following specifications, the absence of reference product specifications or representative samples (QA/QC programme).

Development, application and standardization of quality checking for waste packages must identify R&D requirements and coordinate the development of new test methods.

For HLW glass, further investigation of the secondary phase formation is needed since it could affect the glass dissolution mechanisms in the long-term.

Investigations on the soluble concentrations of the actinides in clay media in the presence of bitumen degradation products, on the ageing behaviour of bitumenized waste and on the microbiological degradation of bitumen due to nitrate and sulphate reducing bacteria are scheduled. Microbiological activity could lead to the production of methane and complexing agents which need to be further studied as gas and chemical effects.

A new laboratory corrosion programme (steel) will be run to provide the necessary information in both aerobic and anaerobic environments.

Chemical stability of Synroc, a titanate ceramic proposed as an alternative to glass, is under evaluation for Boom clay disposal environments.

New programmes on other waste forms, such as cemented reprocessing sludge (BR2/Dounreay), cellulose containing MOX waste, and spent fuel have to be started.

3.4.1.2 Geomechanical/Geochemical Behaviour of Clay-Based Materials

The mechanical and chemical stability of the near field provide a set of controlling factors for groundwater movement, radionuclide transport and heat dissipation through the interface between near and far field. The excavation disturbed zone in clay and the backfill/sealing material might substantially affect the subsequent migration behaviour of radionuclides.

Background

Excavation disturbed zone. The measurements of pore water pressure and stress distribution around boreholes and galleries during and after their construction (Neerdael and De Bruyn, 1988) have shown that the extent of the disturbance depends on the excavation diameter, the excavation speed and the oversizing with respect to the lining diameter; the magnitude of the disturbance drops significantly with distance and decreases slowly with time thanks to the plasticity of clay.

Backfill and sealing material. In case of a repository in clay, a clay-based material is chosen as backfill material for the same reasons as put forward when dealing with the geological barrier. Boom clay has been characterized with a view to reusing the excavated clay spoils for backfill, since oxidation (pyrite) can be prevented easily by fast drying. The following results have been found for highly compacted Boom clay with regard to compacted bentonite: (a) slightly higher but sufficiently low hydraulic conductivity, (b) lower swelling but still convenient regarding the lithostatic pressures at the Mol site, (c) better retention properties for some radionuclides, and (d) steam effect less dramatic than for some bentonites.

When dry compacted clay is used as a backfilling around HLW packages, its behaviour during saturation is complicated by the thermal transient caused by the heat emission from the waste. It must be shown that during this transient period, no (negative) influence on the long term behaviour of the repository can occur due to e.g. shrinkage or collapse of the clay backfill. Therefore, the thermo-hydro-mechanical (THM) and geochemical behaviour of the backfill must be studied. Typically hydro-mechanical and thermo-hydraulical models have to be coupled to describe stress/strain behaviour, moisture migration and heat transfer. The thermo-hydraulic model also has to be coupled to a geochemical code to describe the migration and formation of chemical species.

To avoid the effect that the natural occurring, or human induced, disturbance in one disposal gallery would have on the entire repository, the disposal galleries need to be properly sealed from one another. The shafts must also be properly sealed because they might act as preferential pathways to the biosphere, short-circuiting the Boom clay formation.

The main requirements for a sealing material are: low hydraulic conductivity and long term mechanical and chemical stability. Clay is also the more preferable option in this case. With high density bentonite, the hydraulic conductivity criterion can easily be fulfilled and with a proper composition, the stability requirements can also be fulfilled.

Results

Excavation disturbed zone. Small changes in mechanical stress and pore water pressure have only a minor influence on the migration properties of the Boom clay. Large changes in stress and pore water pressure are limited to the zone close to the gallery or borehole wall.

Backfill and sealing material. A fairly extended database concerning THM properties of unsaturated clays is a prerequisite to enable physical model testing, model calibration and validation. Impressive work has been done on this subject during the last four years in the framework of three research contracts of the present E.C. programme (Volckaert et al., 1994). The results obtained confirm that swelling clays are promising backfill materials.

Future actions

Potential influence of the stress changes on the migration parameters might be studied in combination with temperature, radiation and gas effects.

Quantification of the disturbed zone around a disposal gallery successively submitted to decompression and thermal loading is one of the main objectives of the PRACLAY demonstration test (see Section 3.3.4).

Efforts on the THM behaviour of dense unsaturated clay are continuing for the next five years. A large scale *in situ* demonstration test has been proposed within the next E.C. five year programme.

3.4.1.3 Gas Generation and Transport

In a radioactive waste repository, gas generation may occur due to several phenomena depending on a number of factors (waste types, packages, buffer/backfill, near field host rock). The gas release may increase the rates of flow of potentially contaminated groundwaters to the surface and two-phase flow may entrain radionuclides in solution at the gas-water interface.

Background

The main potential sources of gases in the case of deep geological disposal are methane from degradation of organic materials and hydrogen from anaerobic corrosion of iron. Of these gases, hydrogen is certainly the gas which can be released in potentially the largest amounts.

The objectives of the MEGAS (Modelling and Experiments on GAS migration in repository host rocks) project coordinated by SCK•CEN extend from the understanding of the consequences of gas generation to the validation, using a 3-D *in situ* gas injection experiment, of a gas migration model (TOPAZ-INTERA). Attention has to be paid to gas to ensure that the potential pressure build-up in the near-field of a repository would not lead to fractures in the host medium, which might act as new migration pathways affecting the long term safety of the whole disposal system.

Results

In the surface laboratory, special odometers were built to saturate clay cores to measure its hydraulic conductivity and to perform gas injection; at the end of the gas flow experiment, the cores were still more than 95% saturated, as confirmed by X-ray tomographic analysis. The formation of preferential pathways and a low breakthrough pressure were observed during the field test. These observations are a consequence of the stress distribution around the test borehole. As explained in section 3.4.1.2 (excavated disturbed zone), the drilling of a borehole significantly reduces the stress close to the borehole wall and particularly the effective stress, i.e., the pressure keeping the clay particles together. As a consequence, it is relatively easy to create gas pathways in the excavation disturbed zone. Further *in situ* tests have shown that these fissures close very soon after the gas injection is stopped (Volckaert, et al., 1995).

Future actions

More experimental data are necessary to quantify the link between hydraulic and geomechanical parameters of Boom clay and further gas migration experiments are needed; the use of a swelling backfill material, which locally increases the lower effective stress distribution, would reduce the concentration of pathways in the excavated disturbed zone. Tests in the surface laboratory on larger clay cores will also contribute to increase confidence in the verification and validation of the model.

3.4.1.4 Thermal, Radiation and Chemical Effects

Radiolysis of water by gamma rays can lead to the production of oxidants which might seriously alter the near field chemistry and retention properties of the clay. The radiation effects need to be combined with heat effects, as they occur during the same period. Chemical species released from the waste might seriously alter the near-field chemistry and retention properties of the clay.

Background

The study of the combined effect of heat and radiation is one of the main objectives of the CERBERUS *in situ* experiment (Noynaert et al., 1992). The CERBERUS experiment simulates the in-floor configuration for the disposal of a COGEMA HLW canister after 50 years cooling. Instruments, or test specimens, have been installed in the periphery to obtain data on water chemistry, radionuclide migration, corrosion, radiolysis, water pressure and temperature distribution.

The thermal transient period will last about 300 to 500 years in the case of HLW and 3,000 to 5,000 years in the case of spent fuel. The formation of steam must be avoided at all times because it would lead to operational problems, enhanced corrosion, and could have a dramatic effect on the hydraulic and mechanical properties of clay backfill and sealing materials. A higher temperature will further increase the solubility of the waste form and radionuclides and might reduce the sorption capacity of the clay and/or increase the diffusion coefficients. These effects have not yet been quantified. The temperature cycle also has implications on the mechanical behaviour of the clay. The combined effect of all these influences on the migration in the near field is very complex to model; currently it can not be done with confidence due to a lack of knowledge.

Chemical effects have not yet been studied in detail; however some potential problems have already been identified. The presence of large quantities of degradable organic material and soluble salts, such as nitrates and sulphates, can have an important effect on the global near-field chemistry, the solubility of the radionuclides, and the sorption properties of the clay. The decomposition of e.g. cellulose could generate important quantities of complexing agents.

Results

Seven years of follow-up measurements in the clay around the CERBERUS experiment show a decrease in pH with about one pH unit and a very slight increase in redox potential so that the clay remained reducing. The presence of thiosulphates, which can seriously increase metal corrosion, and oxalate, which can act as a complexant, was observed (Put et al., 1993). The codes for the calculation of the thermal, hydraulical and radiation field were validated.

Selecting the appropriate concept, cooling time and overpack design allows one to avoid an investigation of high temperature and steam effects.

Future actions

Radiation effects will be further studied in the last phase of the CERBERUS experiment, together with the performance of a potential backfill material (Boom clay based) that has been submitted for more than five years to realistic temperature and radiation fields.

Chemical effects identified need further investigation, surely when elaborating acceptance criteria for geological disposal systems. Irreversible complexation of actinides by small organic molecules would strongly increase their transport through the clay. Nitrates affect the redox conditions in the clay while high salt concentrations also influence its sorption properties.

3.4.1.5 Radionuclide Migration

Research on radionuclide migration through the Boom clay has provided data to support the performance assessment of geological waste disposal. Work was concentrated on methodological aspects, codes for flow and transport, theoretical studies, laboratory and field experiments on geochemical transport phenomena to develop and test conceptual and numerical models.

Background

Radionuclide transport in porous and fissured media is controlled by diffusion, advection, retardation (sorption, precipitation) processes and radioactive decay, as described in a general way by the advection-dispersion equation. In a rather stiff material, such as Boom clay, the radionuclide migration is dominated by diffusion. Advection plays a secondary role due to the very low hydraulic conductivity of this medium and the absence of preferential paths for the water (self-closure of fissures due to the clay plasticity).

Boom clay exhibits good sorption properties for cations due to its physico-chemical characteristics under *in situ* conditions. The presence of micro-dispersed pyrite and organic matter intimately associated with the clay mineral surface ensures strong reducing conditions while carbonate (calcite and siderite) maintains a slightly alkaline environment. These physico-chemical conditions are very favourable for the sorption of cations on the Boom clay surface.

From the performance assessment studies, three groups of radionuclides have been selected for further experiments in the present migration program:

- critical radionuclides: ^{14}C,^{99}Tc,^{129}I,^{135}Cs and ^{237}Np;
- less critical radionuclides: ^{79}Se, ^{93}Zr, ^{107}Pd, U-, Pu-, Am-, Cm-isotopes; and
- non-critical radionuclides used for the study of migration mechanisms in Boom clay (Sr^{2+}, Br^{-1}, HTO) are used as chemical analogues of relevant nuclides (Eu^{3+} as an analogue for the actinides).

The migration behaviour of radionuclides in the Boom clay is studied by means of different types of experiments (De Preter et al., 1992):

- fundamental physico-chemical studies of radionuclides in the clay;
- laboratory experiments for the measurement on clay cores of the migration (and hydraulic) parameters (diffusion accessible porosity, retardation factor, diffusion coefficient etc.); and
- large scale *in situ* experiments for model validation and confidence building.

Mathematical models have been developed for the design and interpretation of migration experiments performed both on small scale samples and *in situ* at a larger scale. Different analytical models taking into account the various kind of experiments, the type of source terms and the boundary conditions are presently available to interpret laboratory migration experiments and to determine the migration parameters of the radionuclides. The tridimensional migration model and the values of parameters obtained in the surface laboratory on small clay plugs (3 cm) for HTO (tritiated water) have been validated on the metric scale (2 m) by comparison with model predictions from experimental results with large scale *in situ* injection of tritiated water.

Results and future actions

Running studies confirm that the most critical elements to be considered for long term safety are iodine and fission products.

Laboratory experiments have to be performed under strictly anaerobic conditions. Asmall exposure to traces of oxygen alters the physico-chemical characteristics of Boom clay; many radionuclides (Tc, U, Se, etc.) are very sensitive to oxidation. In the case of technetium e.g., the strongly sorbed $Tc0^{2+}$ cation is rapidly converted to the non-sorbed $Tc0^{4-}$ anion if the oxidation potential of the interstitial solution increases too much.

Aqueous speciation, the clay surface properties, and the concept of anion exclusion or diffusion accessible porosity are fundamental in the migration of radionuclides.

Organic matter distributed between the solid phase (99.99%) and the interstitial solution (0.01%) plays a dual role in the sorption and the complexation processes of the trivalent actinides and lanthanides in Boom clay.The coating of organic matter at the Boom clay surface increases the surface complexation properties of the solid phase and enhances the retention of trivalent radionuclides on the clay. On the contrary, small dissolved organic molecules might form soluble complexes with these nuclides increasing their mobility in water.

3.4.2 Demonstration Tests in Underground Research Facility.

The HADES underground research facility is presently the only one installed in a clay layer. The facility offers important opportunities for participation of foreign organisations and contributes to the scientific leadership of SCK•CEN for waste disposal in clay.

Objectives

Investigate and demonstrate the feasibility of disposal concepts for vitrified high level waste, long-lived waste and spent fuel and provide reliable data on the performance of repository barrier components; and allow testing and validation of models under representative conditions.

Research topics

The research carried out at SCK•CEN is related to the following scientific and technical aspects:

- testing and demonstration of disposal concepts;
- backfilling and sealing a repository;
- long term behaviour of repository components;
- ground water flow analysis and radionuclide migration; and
- monitoring the reconsolidation of the disturbed zone around an excavation.

All of these items were already introduced in Section 3.4.1. The corresponding demonstration tests now (or previously) running are briefly described below.

In disposal holes (in-floor concept), for demonstration purposes:

- a large scale integrated experiment simulating the radiation and thermal output of a COGEMA canister after 50 years cooling time (CERBERUS). Different experimental set-ups are installed in the periphery to get data on corrosion, migration, radiolysis and material performance; and
- full scale thermo-hydro-mechanical experiments under similar thermal conditions but according to other designs and instrumentation purposes and considering the use of several potential backfill materials (BACCHUS 1&2, CACTUS 1&2).

At the horizontal (in-gallery or axial concept), for validation purposes:

- large scale thermo-hydro-mechanical experiment (ATLAS), again under similar thermal conditions but without the use of backfill material between the clay wall and the heaters;
- 3-D gas migration test (MEGAS) using in-house designed injection and detection piezometers; and
- 2-D and 3-D radionuclide migration experiments using non-sorbed tracers (tritiated water, etc.)

The results of all these tests confirm Boom clay as a promising host rock for radwaste disposal.

In the framework of the fourth R&D programme on “management and storage of waste” of the E.C., we also developed two experiments for the French waste management authority (ANDRA):

- ARCHIMEDES project, now running for three years, aims at studying the acquisition and regulation of the water chemistry in a clay environment. The work programme consists of the following four main topics: (a) fluid and solid sampling and characterization, (b) *in situ* measurements, (c) microbial interactions; and (d) fluid-rock interaction modelling. Available results clearly show, e.g., that the microbial activity in the far field is at the limit of detection while the near-field exhibits the same microbial activity as a soil; and
- PHEBUS project aims at understanding the phenomenology of hydrical exchanges between ventilated underground structures (operational phase) and the host clay.After support tests and mock-ups in surface laboratories, an *in situ* ventilation experiment has been installed from the URF and has been operational since May 1995.

An important action to be developed for the next ten years is the PRACLAY project. PRACLAY is a demonstration test simulating the thermal output of a 30 m long HLW disposal gallery, 2 m in diameter (see Section 3.3.4). The experiment will be installed from an extension of the existing facilities to be built over the next 3 years. The experiment is planned to last until 2005.

3.4.3 Safety Aspects of Waste Disposal

The tools needed for a preliminary evaluation of the safety of disposal of radioactive waste in Boom clay have been described. Performance assessment is a phased process and the models and data used have to be adapted as results from the R&D programme become available.

This can be illustrated by the study on regional hydrology, which is above all one of the support research items for performance assessments. In the period 1981-1984, a regional site characterisation programme has been carried out. It consisted mainly of drilling about 20 boreholes located in northeast Belgium and the elaboration of pumping and slug tests to determine transmissivities and hydraulic conductivities in the aquifers. These boreholes have been equipped as piezometers and together with existing piezometers they form a regional piezometric network that consists of 122 observation points. The water levels in the piezometers have been measured monthly since 1984. However, most piezometers are located in the overlying Neogene aquifer. The scarcity of data from the underlying Ruisbroek-Berg aquifer is a considerable drawback for the hydrogeological modelling of the regional aquifer system and urgently needs remedial action to enable improvement of the available models.

Objectives

Develop and apply a comprehensive approach and methodology for evaluating the long term safety of disposal in clay, including radiological as well as non-radiological (chemical-toxic) consequences for the purpose of increasing confidence in building and public acceptance by putting the long term risks into perspective.

Background

Aperformance assessment of a geological repository for radioactive waste disposal consists of three steps:

1. identification of scenarios that can lead to exposure of man and an estimation of the probability of occurrence of these scenarios;
2. evaluation of the radiological consequences of the identified scenarios; and
3. comparison of the estimated doses and risks using appropriate radiological protection criteria.

Since no criteria or guidelines applicable to radioactive waste repositories have been published in Belgium, the recommendations of the International Commission on Radiological Protection (ICRP) are used as a reference. The ICRP recommends the application of a total dose limit to individuals from all relevant sources. The annual effective dose limits are 1 mSv/a for a member of the public and 20 mSv/a for occupational exposure. These limits are considered by ICRP as the boundary between "unacceptable" and "tolerable". Concerning potential exposures, ICRP recommends that risk limits be of the same order of magnitude as the risks corresponding to the dose limits for routine exposure.

In order to meet the requirements of individual dose or risk limits for all relevant sources, ICRP proposes to determine dose or risk constraints related to sources. In several European countries, e.g. Switzerland, the dose constraint has been taken equal to 1/10 of the annual dose limit for the case of geological repositories. We recommend using the same value in Belgium. This dose constraint of 10^{-4} Sv/a is clearly indicated in the diagrams reproduced in this section. The corresponding risk constraint is of the order of $5x10^{-6}$/a for fatal radiation induced cancers.

Adifficult aspect of performance assessment is the time scales that have to be considered. In France, the radiological protection authority requires forecasts of doses based on an explicit uncertainty analysis for the first 10,000 years. For a period of 100,000 years, the impact of expected changes in climate on the repository system have to be taken into account. For very long time scales, "more qualitative assessments have to be made to verify that the release of the radionuclides does not result in an unacceptable individual dose."

Results and future actions

Hydrogeological modelling. A new version of the regional hydrogeological model for the Mol site (Wemaere and Marivoet, 1995) has been achieved by considering the available data and computer techniques and after reviewing all the improvements that could be brought to the model developed in 1985. A detailed geometry is used to reproduce the geological structure in the model, and special attention is given to the fault system in the northeastern part of the modeled area. The calibration of the regional model, which was strongly hindered by the poor characterization of the underlying aquifers, led to a value of $3x10^{-10}$ m/s for the hydraulic conductivity of the Boom clay on a regional scale. This means that the calibrated value is much higher than the values measured *in situ* or on cores taken from the underground laboratory ($3x10^{-12}$ m/s). This discrepancy should be further investigated. The latter value is used for the clay migration model in the performance assessment, because in that model a local value of the hydraulic conductivity for clay is needed.

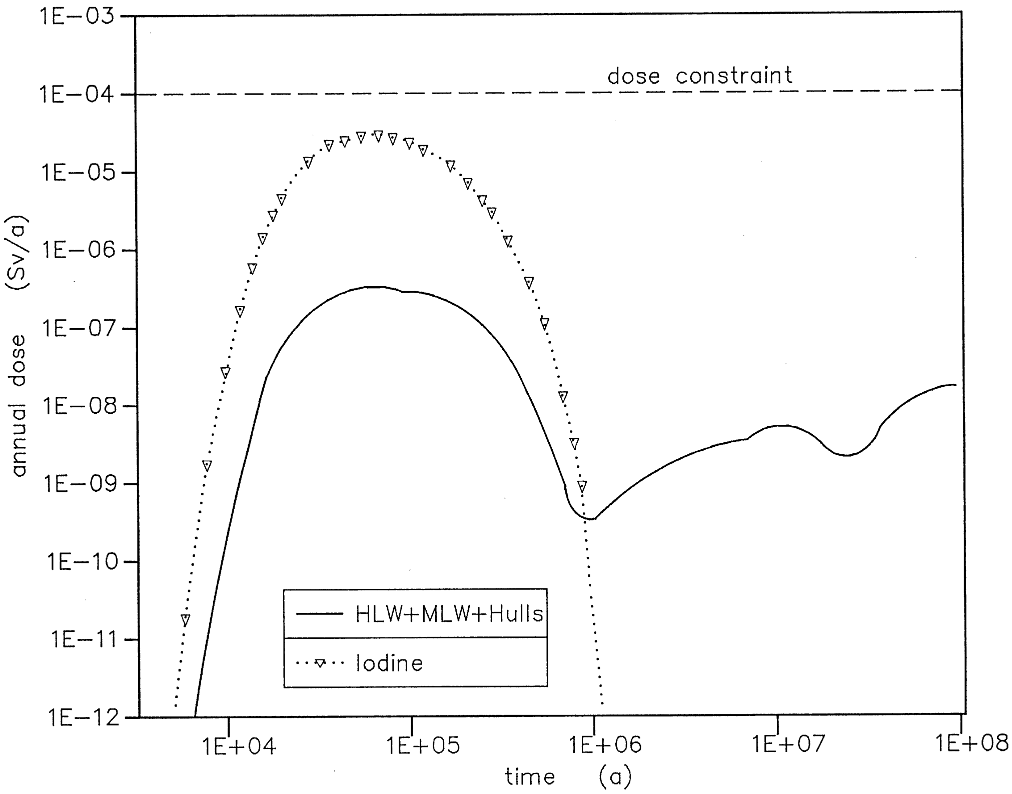

Figure 3.4. Normal evolution scenario - Annual doses via the water well pathway.

Additional hydrogeological investigations focusing on the spatial variability of the parameters of the Boom clay and on a better characterization of the aquifers, especially the deepest ones, underlying the Boom clay are highly recommended. Further, the hydrochemistry should help to understand the behaviour of the aquifer system and constrain its modelling since stable isotope analysis and major trace element analyses provide a way to understand the origins of the water and its evolution.

Performance assessments of re p rocessing waste. Following the CEC's PAGIS and PACOMAstudies, the "Updating 1990" study (Marivoet, 1992) was performed. A normal evolution scenario, which considers all phenomena that are certain or highly likely to occur, as well as some altered evolution scenarios (deep water well, climatic change, tectonic fault) have been identified in this way. Because the Neogene aquifer is an important drinking water resource, the pumping of drinking and irrigation waters from a well located in that aquifer is considered as a realistic pathway to the biosphere within the normal evolution scenario. The consequences calculated in the case of this scenario for various considered waste types are shown in Figure 3.4.

For the reference waste types (HLW, hulls and MLW) together, the highest calculated annual doses, which are only $3x10^{-7}$ Sv/a, are due to the small fractions (about 1%) of I-129 present in the hulls and the medium-level waste.

The deterministic performance calculations are complemented with stochastic calculations in which the uncertainties in the model parameters are taken into account. The results of such calculations show that the expected value for the annual dose and the 95% percentiles also remain below the dose constraint. Figure 3.4 also indicates that a possibly problematic waste type consists of the iodine waste. This waste type is not included on the list of waste types that have to be considered in Belgium (NIRAS, 1989). This waste arises in the reprocessing plants from the capture of iodine from the dissolver gasses and contains 98% of the I-129 which is generated by nuclear fission. Since iodine is not sorbed on the Boom clay, the disposal of this waste form might, in case all of it were returned to Belgium, lead to some scenarios for annual doses of the order of the dose constraint.

In 1991, the CEC's EVEREST project was started.

Complementary sensitivity studies are investigated within EVEREST to determine which elements of the repository system strongly influence the performance of the repository system. Stochastic calculations were performed to assess the influence of the uncertainties in the model parameters on the calculated annual doses. Other sensitivity studies focused on conceptual model uncertainty in the aquifer modelling and on uncertainties in the scenario descriptions.

In 1992, the next Belgian performance assessment, ordered by NIRAS/ONDRAF, started with the description of a systematic scenario study. Recently two additional scenarios have been analysed (Wibin and Marivoet, 1995). The exploitation scenario considers the pumping of water from the Ruisbroek-Berg aquifer which underlies the Boom clay layer. The consequences associated with this scenario are more than two orders of magnitude higher than the doses estimated for the normal evolution scenario in which only pumping from the overlying aquifer was considered. The “poor sealing” scenario considers that the sealing of the transport gallery and the access shaft are not successful. The consequences of this scenario are not much higher than the ones estimated for the normal evolution scenario.

The next step will be a detailed analysis of the normal evolution scenario. In this analysis, all phenomena which are about certain to occur will be considered. Thus, the expected evolution of the climate for the next 100,000 years has to be taken into account. The impact of the climate changes on the aquifer system will be evaluated by applying geoprospective methods. In this time period, two glaciations are expected to occur that will result in drastic changes in the geography and shallow geology. Consequently, the uncertainties in the aquifer system become considerable for these time scales. However, it should be noticed that these climate changes will have very little influence on the migration of radionuclides in the Boom clay, which is about 32 million years old and would remain stable for another million years.

The case of spent fuel. SCK•CEN started a preliminary performance assessment of the direct disposal of spent fuel in a clay layer (Marivoet et al., 1995). The spent fuel types considered are uranium oxide fuels with burn-ups of 33 and 45 MWd/tHM and MOX fuel with a burn-up of 45 MWd/tHM. The radionuclide inventories have been calculated with the ORIGEN2 code. A serious problem in the assessment is that no information is available from specific experiments on the behaviour of

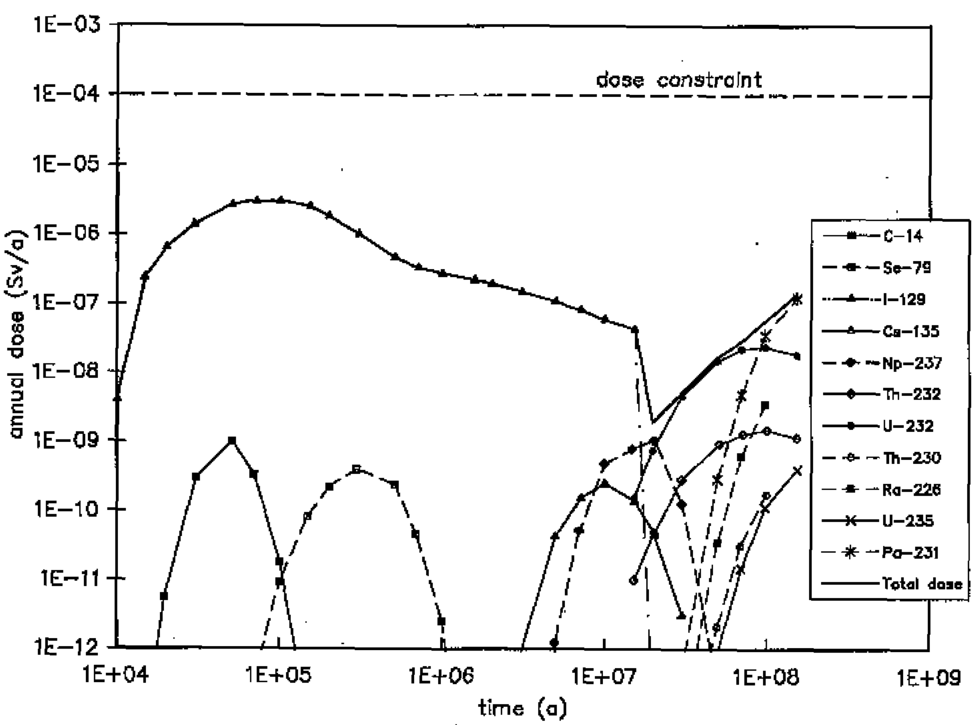

Figure 3.5. Preliminary performance assessment of the direct disposal of spent fuel in clay. Calculated annual doses via the water well pathway (normal evolution scenario).

spent fuel in conditions similar to the ones prevailing in a repository in clay. Therefore, the near-field model is largely based on results obtained from research programmes (mainly Canadian and Swedish), on the disposal of spent fuel in granite.

The annual doses calculated in the case of the normal evolution scenario are given in Figure 3.5. The maximum calculated annual dose is due to I-129 and should occur about 60,000 years after disposal. The maximum annual dose actually depends on the fraction of iodine which is present in the gap between pellets and zircalloy hulls (about 10%). The bulk of the iodine (90%) is dissolved in the uranium oxide crystals and is released over a 10 million year period. A second but lower dose peak due to actinides is expected to occur after a very long time period (several tens of millions of years). The preliminary assessment shows that, from a radiological point of view, the direct disposal of spent fuel in a clay layer is an acceptable option.

Chemical toxicity aspects in the R&D geological disposal programme. A preliminary assessment of the chemical toxicity aspects, related to the geological disposal of radioactive waste, has been carried out. Anumber of simplifying assumptions have been made for this assessment, and the study considered only the possible toxicity of heavy and other metals present in the waste forms or the packing materials. The elements that caused the highest concentrations in the overlying aquifer were uranium, molybdenum and nickel. However these concentrations are lower than the concentration limits given in a European guideline for water to be used for drinking water purposes. The calculated low concentrations can be attributed to the low solubilities of some metals and the fact that most of the expected species in solution are cations, which are strongly sorbed on the Boom clay.

However, this assessment is still preliminary. For many essential parameters like solubility limits and retention coefficients, rough estimates had to be used in the simulations because no experimental data were available. The consequences of beryllium, which arises from the beryllium matrices of SCK•CEN's research reactor BR2, were not analysed because beryllium is essentially toxic to the lungs; therefore, the analysis requires the application of a detailed biosphere model to assess the consequences of the inhalation of beryllium.

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